

Appln. No. 10/777,229
Reply to the Office Action of November 30, 2005

REMARKS

Claims 48, 49, 56-58, 116 and 117 have been canceled. Claims 46, 47, 50-55, 59-117 and new Claims 118-120 are active in the case. Claims 71-115 stand withdrawn from consideration. Reconsideration is respectfully requested.

Drawing Amendments

In compliance with the Draftsperson's review of the drawings as set forth on Form 948, applicants hereby submit replacement drawings. The replacement drawings do not introduce new matter into the case.

Claim Amendments

Claim has been amended to recite that the catalytic composition is comprised of (i) gallium, (ii) at least one lanthanide element, and (iii) a zeolite selected from the MFI family, the crystal lattice of which is made-up of silicon oxide and aluminum oxide in a molar ratio of > 20 to < 500, and is comprised of crystallites, at least 90 % of which have diameters smaller than 500 Å. Support for the claimed molar ratio range can be found in canceled Claim 49. Other dependent claims have been amended in order to make minor corrections thereto.

Support for new Claims 118 and 119 can be found on pages 12 and 13 of the specification and support for Claim 120 can be found in canceled Claim 116. Entry of the amendments to the claims and the new claims is respectfully requested.

Claim Rejection, 35 USC 112

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As to the matter of the one or more aliphatic hydrocarbon compounds that are used as the feed material in the present process, applicants believe that it is not necessary to recite in the claim how the one or more hydrocarbon compounds are produced. Any source of these one or more compounds can be used, and these compounds are readily available in the market place. Claim 46 simply is not dependent on how the hydrocarbon feed is made. Withdrawal of the issue is respectfully requested.

As to the matter of Claims 53, 54 and 56, since Claim 46 has been limited to the family of MFI zeolites, there is no ambiguity in these claims regarding what type of zeolite is employed.

The amendments made to Claim 63 are believed sufficient to obviate the issue that has been raised. Withdrawal of the non-reference ground of rejection is respectfully requested.

Abstract of the Disclosure

The application has been provided with a new abstract as requested.

Invention

The present invention, as now defined, is directed to a process for the production of aromatic hydrocarbon compounds which comprises contacting one or more aliphatic hydrocarbons containing from 3 to 6 carbon atoms with a catalytic composition comprising (i) gallium, (ii) at least one lanthanide element, and (iii) a zeolite selected from the MFI family of zeolites, the crystal lattice of which consists of silicon oxide and aluminum oxide having a molar ratio ranging from > 20 to < 500. The zeolite is comprised of crystallites, at least 90 %

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of which have diameters smaller than 500 Å.

Claims 46-51, 53-70, 116 and 117 stand rejected based on 35 USC 103(a) as obvious over Chu, U. S. Patent 4,891,463 in view of Hirandi et al, U. S. Patent 4,854,939. This ground of rejection is respectfully requested.

The technology of the Chu patent is clearly related to the present invention since it discloses a process of producing aromatic hydrocarbon compounds by contacting a hydrocarbon feed in which the hydrocarbon compounds contain from 2 to 12 carbon atoms with an acidic crystalline gallosilicate zeolite catalyst in which some portion of the gallium is incorporated in the zeolite structure in tetrahedral form. However, a significant difference between the zeolite catalyst of the present process and that of the reference is that while the patent teaches a SiO_2 to Al_2O_3 molar ratio for the zeolite of at least about 12 (col 3, line 60), the preferred SiO_2 to Al_2O_3 molar ratio range for the zeolite component in the patent is 500 to 26,000. Contrast this range with the SiO_2 to Al_2O_3 molar ratio range for the zeolite of the present claims of > 20 to < 500. In fact, the patent at column 4, lines 25-26 teaches a minimum molar ratio of 550 or at least 880 or more at column 4, lines 13-14. Exemplified catalysts in the patent employ zeolites of a SiO_2 to Al_2O_3 molar ratio of 880 and 850 and comparative examples employ zeolites having a SiO_2 to Al_2O_3 molar ratio of 70 and 40. Thus, it can be said that the zeolite component of the present catalyst system is different from the zeolite component that the patent would lead one of skill in the art to employ in the subject hydrocarbon conversion reaction.

Another clear distinction between the catalyst system of the patent and that of the present invention is that a lanthanide element is an essential component of the present catalyst

system, whereas this is not the case in the patent. The patent at column 3, lines 35-54 discloses the possible addition of certain metals from Groups I through VIII of the Periodic Table to the zeolite and also mentions rare earth metals at line 54. However, there is no teaching anywhere in the patent of the selection and use of just one lanthanide element for the modification of a zeolite catalyst that would be advantageous in some manner for the conversion reaction. Moreover, there is no teaching or suggestion of the use of rhenium as a component of a zeolite catalyst for the disclosed hydrocarbon conversion reaction.

It also should be noted that the patent teaches that the gallium component is present in the framework of the zeolite (col 2, lines 17-19; 37-38, 47-48, 53-55, 57-58 and 64-65; col 3, lines 39-41). Further, the presence of gallium within the zeolitic framework imparts a Bronsted acidity to the catalyst composition that is weaker than that of a zeolite that contains alumina. (Column 3, lines 43-44). Accordingly, the Chu patent does not show or suggest the invention as claimed.

As to the matter of the Harandi patent, the same teaches a catalyst system for an integrated process in which iso-olefins and methanol are combined to produce methyl tert-alkyl ethers while olefins and perhaps some paraffins are simultaneously converted to aromatic hydrocarbons, the reactions being catalyzed by a zeolite catalyst. The methanol that is formed that also does not react to produce methyl tert-alkyl ether product, co reacts with ethylene and C₃-hydrocarbons. The catalyst employed in this process is ZSM-5. Preferably, the catalyst is in the acid form and has a silica/alumina ratio of 20 to 200, preferably from 30 to 70 and may include metals such as Zn, Cu, Ga and Pt. The ZSM-5 catalyst may have a crystallite size of about 200 to 500 Å.

It is evident from the discussion above that a first major distinction between the present invention and that of Harandi is that feed material to the reaction of the patent is completely different than the hydrocarbon feed of the present invention, but also that of the Chu patent. Harandi employs a feed that contains, in addition to C₃- and C₅-hydrocarbons, high quantities of methanol and ethylene. On the other hand, the feed of the Chu patent contains at least 50 wt % of at least one aliphatic hydrocarbon containing from 2 to 12 carbon atoms. Thus, the feed of Chu is markedly different from that of Harandi et al, while, as can be ascertained from the above discussion, the hydrocarbon feed of Chu is not necessarily the same as that of the present invention.

Another important aspect by which the invention of Harandi et al also differs from that of Chu and the present invention is in the catalyst employed. Harandi et al employs a zeolite of the ZSM-5 type, preferably microcrystalline, which may contain an additional metal such as Zn, Cu, Ga and Pt. None of these metals are claimed or exemplified. Further, there is no disclosure as to effective quantities of these metals. There is no hint in the reference that Ga is in some preferred as a catalyst component. Moreover, there is no hint as to the important lanthanide element component of the present zeolite based catalyst system. Thus, the combined prior art does not suggest the present catalyst system.

It has been mentioned above that Harandi et al refers to a microcrystallinity of the ZSM-5 zeolite. However, no mention of what this specifically means is presented in the patent. There is a disclosure at column 5, lines 36-39 of the patent which states that the zeolites have a crystalline size ranging from 0.02 to 0.05 μm , but no mention is made if such a size refers to the diameter or to the radius of the microcrystals. This lack of specifics is significant in view

of the fact that different methods can be used to determine microcrystallite size can be used, and further, different meanings can be deduced according to the method employed. Examples are: (i) the average diameter may be calculated by the Sherrer equation and the distribution of crystallite sizes may be detected by micrographs TEM. Because the Harandi et al patent does not specify the method and therefore the meaning of the term “microcrystallite size” that is used, the patent does not teach a meaningful morphologic parameter for the catalyst. Consider the fact that in Examples 20, 22 and 24 of the present application, the average diameters of 280, 245 and 505 Å are disclosed. These values all could fall within the scope of the definition of microcrystallite size in Harandi et al. However, the present invention makes a morphologic distinction in that the morphologic parameter is defined on the basis of a distribution of crystallite sizes such that 90 % of the crystallites must be within a size range of less than 500 Å in diameter. It therefore appears that there is no way to correlate the microcrystallite zeolites of Harandi with not only the zeolite of the present catalyst system, but also the zeolite catalyst system disclosed in Chu. Even if the comments above about the crystallinity of the zeolites employed by Harandi et al are not considered, there is no reason to believe that a ZSM-5 type of microcrystalline zeolite catalyst of Harandi et al that contains an added Zn, Cu, Ga or Pt metal is suitable as a replacement for the zeolite catalyst employed in the process of Chu in order to achieve better results as a substitute catalyst system for the catalyst system of Chu. There does not appear to be a reasonable basis for combining the two patents as the Examiner has done in the expectation that the process of the present invention would be realized. Rather, applicants submit that a combination of the two patents would suggest a zeolite which must contain gallium in a zeolite framework having a low alumina content, which eventually has

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gallium deposited on the framework of the zeolite, which may contain one element selected from the group of Zn, Pt, Re, Co, Ti, Te, Na, Ni, B, Cr, V, Cu, Pd, Ca and the rare earth metals, and which is comprised of crystals having a size ranging from 200 to 500 Å. The present catalyst system of a NFI zeolite containing both Ga and a lanthanide metal as essential components is not shown or suggested by the combined patents. Withdrawal of the prior art ground of rejection is respectfully requested.

It is now believed that the application is in proper condition for allowance. Early notice to this effect is earnestly solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER AND NEUSTADT, P.C.
Norman F. Oblon



Frederick D. Vastine, Ph.D.
Registration No.: 27,013

Customer Number

22850

(703) 413-3000

(703) 413-2220 (fax)

NFO/FDV